Emissions Control for Lean Gasoline Engines

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  - Calvin Thomas, Dr. Jochen Lauterbach
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- **Collaboration with partners at Umicore:**
  - Davion Clark, David Moser, Chris Owens, Ken Price, Tom Pauly

Project Overview

Timeline

- Year 3 of 3-year program
  - **Project start date:** FY2016
  - **Project end date:** FY2018
- Builds on previous R&D in FY13-FY15

Budget

- FY17: $400k (Task 2*)
- FY18: $400k (Task 2*)

*Task 2: Lean Gasoline Emissions Control

Part of large ORNL project "Enabling Fuel Efficient Engines by Controlling Emissions" (2015 VTO AOP Lab Call)

Barriers Addressed

U.S. DRIVE Advanced Combustion & Emission Control 2018 Roadmap Barriers & Targets:

- U.S. EPA Tier 3 Bin 30 emission standard
- Low-cost, lean-NOx aftertreatment catalysis/system
- Increasing brake engine efficiency

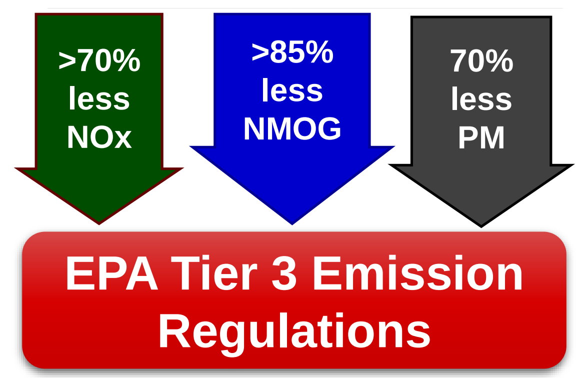
Collaborators & Partners

- General Motors
- Umicore
- University of South Carolina
- Cross-Cut Lean Exhaust Emissions Reduction Simulations (CLEERS)

Objectives and Relevance

Enabling lean-gasoline vehicles to meet emissions regulations will achieve significant reduction in petroleum use

- Objective:
  - Demonstrate technical path to emission compliance that would allow the implementation of lean gasoline vehicles in the U.S. market.
    - Lean vehicles offer 5–15% increased efficiency over stoichiometric-operated gasoline vehicles
    - Compliance required: U.S. EPA Tier 3
  - Investigate strategies for cost-effective compliance
    - minimize precious metal content while maximizing fuel economy

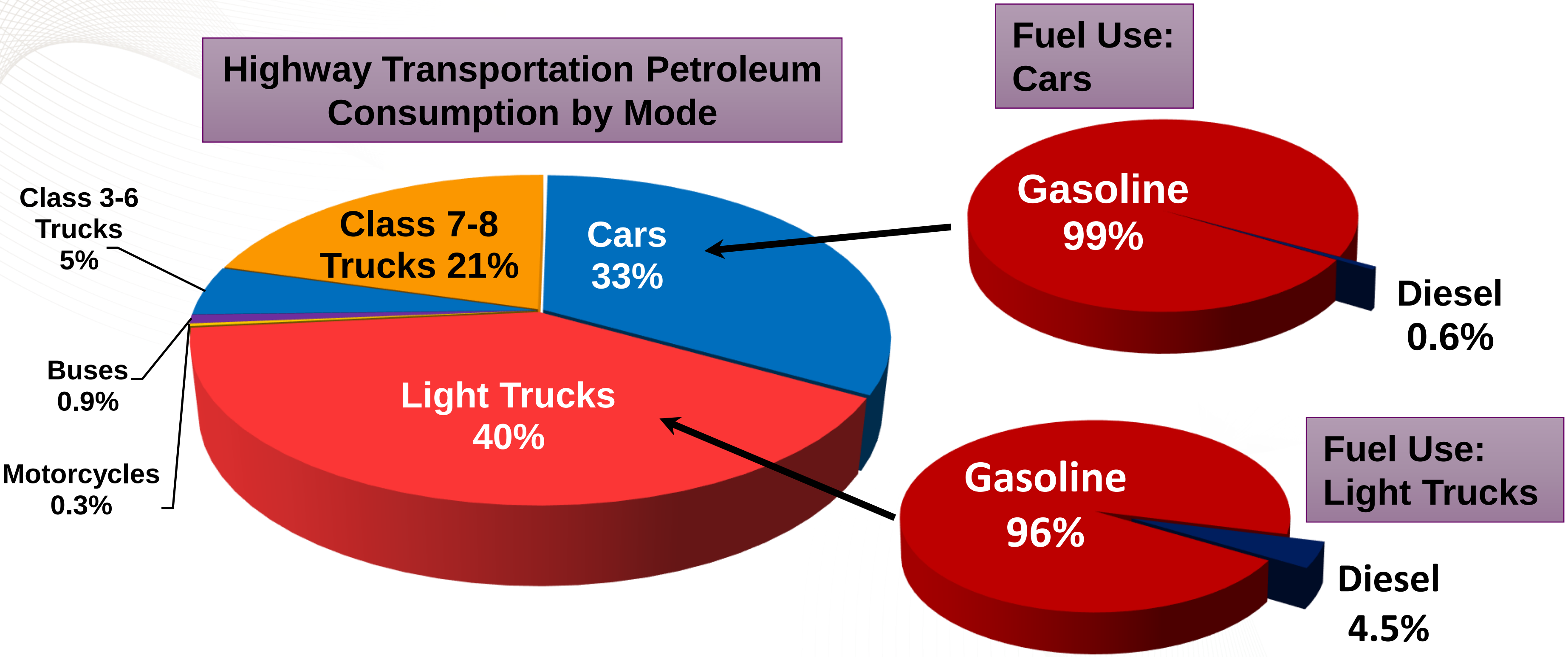


- Relevance:

54.5 mpg CAFE by 2025

  - U.S. passenger car fleet is dominated by gasoline-fueled vehicles.
  - Enabling introduction of more efficient lean gasoline engines can provide significant reductions in overall petroleum use
    - thereby lowering dependence on foreign oil and reducing greenhouse gases

Relevance: small improvements in gasoline fuel economy <u>significantly</u> decreases fuel consumption



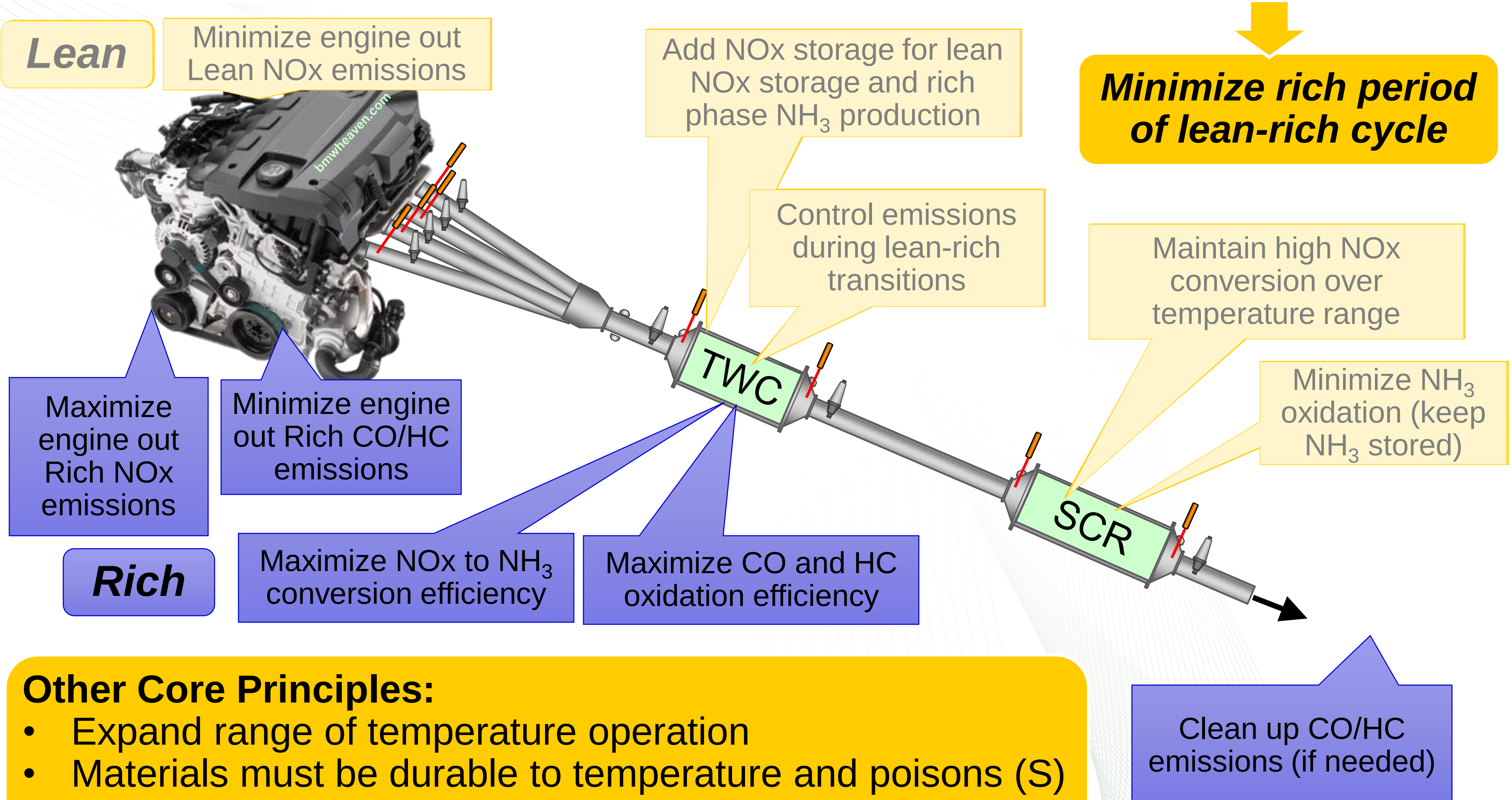
- US car and light-truck fleet dominated by gasoline engines
- 10% fuel economy benefit has significant impact
  - Potential to save 13 billion gallons gasoline annually
- HOWEVER…emissions compliance needed!!!

Lean gasoline vehicles can decrease US gasoline consumption by <u>~13 billion gal/year</u>

Reference: 2016 Transportation Energy Data Book, Ed. 35 (2014 data)

Approach focuses on catalyst and system optimization of Passive SCR (and LNT+SCR)

Key Principle: system fuel efficiency gain depends on optimizing NH_3 production during rich operation and NOx reduction during lean operation



Other Core Principles:

- Expand range of temperature operation
- Materials must be durable to temperature and poisons (S)
- Understand Pt group metals utilization to minimize cost

Approach focuses on catalyst and system optimization of Passive SCR (and LNT+SCR)

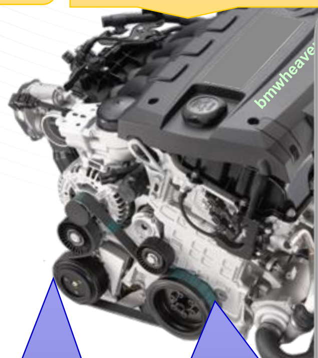
Key Principle: system fuel efficiency gain depends on optimizing NH_3 production during rich operation and NOx reduction during lean operation

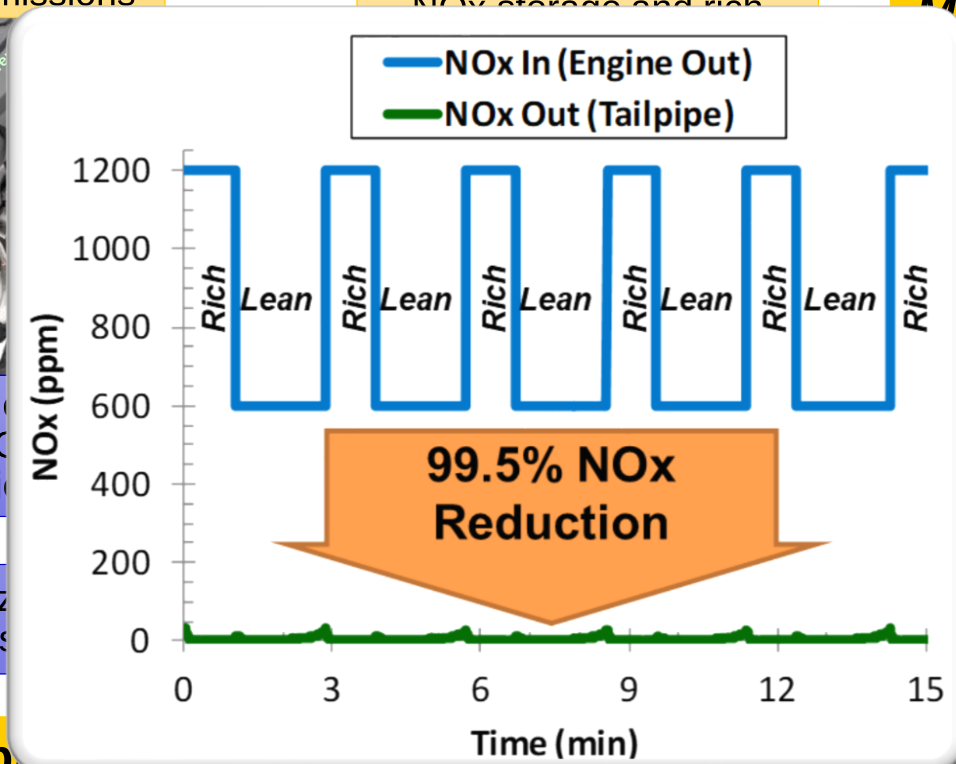
Lean

Minimize engine out Lean NOx emissions

Add NOx storage for lean NOx storage and rich

Minimize rich period of lean-rich cycle





Maintain high NOx conversion over temperature range

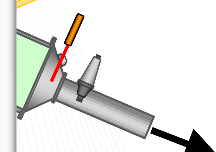
Minimize NH_3 oxidation (keep NH_3 stored)

Maximize engine out Rich NOx emissions

Minimize out Rich C emissi

Rich

Maximiz convers



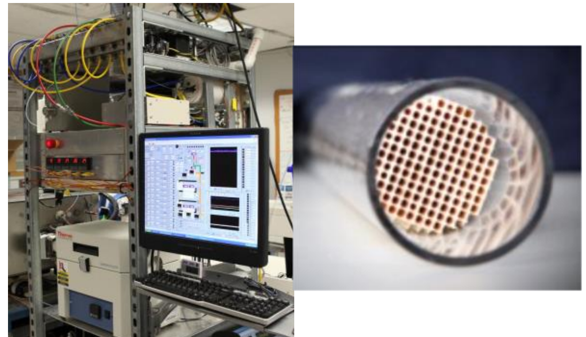
Clean up CO/HC emissions (if needed)

Other Core Principl

- Expand range of temperature operation
- Materials must be durable to temperature and poisons (S)
- Understand Pt group metals utilization to minimize cost

Approach: Iterative Flow Reactor + Engine Study

BMW 120i lean gasoline engine platform with NI open controller



Automated Flow Reactor with feedback control and tandem catalysts

Aging Rig, Automated Flow Reactors, detailed characterization

- Define exhaust conditions
- Measure TWC performance vs. λ
- Quantify TWC+SCR emissions & fuel efficiency
- Optimize combustion parameters and evaluate full system performance

- Identify formulation impacts on TWC performance
- Evaluate SCR formulation effects
- Investigate alternate catalyst configurations, operating strategies

- Age, poison, characterize selected TWCs
- Age, characterize selected SCR(s)

umicore Automotive Catalysts — *Prototype Catalysts & Insights*

GM — *Technical Guidance*

Collaborations with modeling community and CLEERS

Collaborations and Partners

Primary Project Partners

- **GM**
  - guidance and advice on lean gasoline systems via monthly teleconferences
- **Umicore**
  - guidance (via monthly teleconferences) and catalysts for studies (both commercial and prototype formulations)
- **University of South Carolina (Jochen Lauterbach)**
  - Catalyst aging studies with student Calvin Thomas

Additional Collaborators/Partners on Project/Engine Platform (Since Project Inception)

- **CDTi:** catalysts for studies
- **CLEERS:** Share results/data and identify research needs
- **LANL:** Engine platform used for NH_3 sensor study (Mukundan, Brosha, Kreller)
- **MECA:** GPF studies via Work For Others contract
- **University of Minnesota:** Collaboration on DOE funded project at U of Minn. related to lean GDI PM (PI: Will Northrop)
- **CTS (formerly FST-Filter Sensing Technologies):** FOA project on RF sensors for GPF, SCR, TWC on-board diagnostics
- **Tennessee Tech University:** Project data being used for lean gasoline emission control system modeling
- **DOE VTO Fuel Technology Program:** Engine platform used for biofuel-based HC-SCR studies and TWC employed in Co-Optima research

R&D Expanded Coverage via Collaborations:

- Lean GDI PM Control
- Sensors
- Modeling
- Fuels

Milestones

Quarterly Milestones

Complete

- **FY2017, Q3:** Evaluate three commercial or commercial-intent SCR catalyst formulations under dynamic air/fuel ratio operation relevant to lean gasoline engine application.

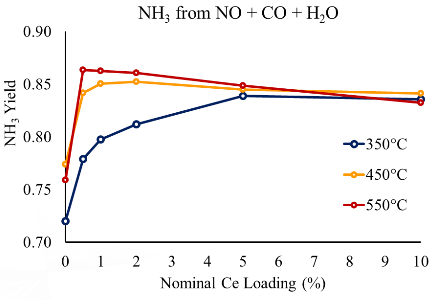
Annual SMART Milestones

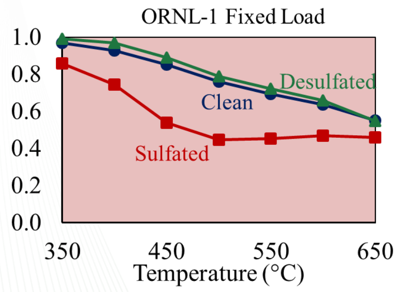
Complete

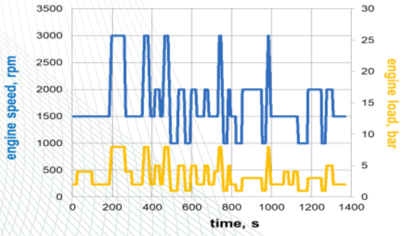
- **FY2017:** (SMART) Meet EPA Tier 3 emission levels with a lean GDI engine while using less than 4 g Platinum Group Metal per liter of engine displacement (cost-related metric) and determine fuel efficiency benefit over USDRIVE naturally aspirated gasoline engine baseline efficiency at eight speed and load points defined by industry collaborators GM and Umicore. Based on drive cycle modes, determine which speed and load points are feasible for lean operation.

Summary of Technical Accomplishments

- Full Review of project at USCAR in August 2017
  - Attended by representatives of GM, Ford, and FCA
  - Reviewed over 5 years of research and provided future directions in detail for feedback
- Evaluated impact of Ce loading on NH_3 production over model TWC catalyst formulations
  - As little as 0.5% Ce significantly promotes NH_3 formation
- Completed sulfur sensitivity analysis on two TWCs
  - Used probe reactions to evaluate lost functionality
  - NH_3 production remains high on NSR-TWC
- Using two catalyst system met Tier 3 NOx+HC (0.03 g/mi) with 5.9% fuel efficiency improvement
  - NH_3 and CO issues, but points to specific improvements needed in strategy and emissions control catalysts





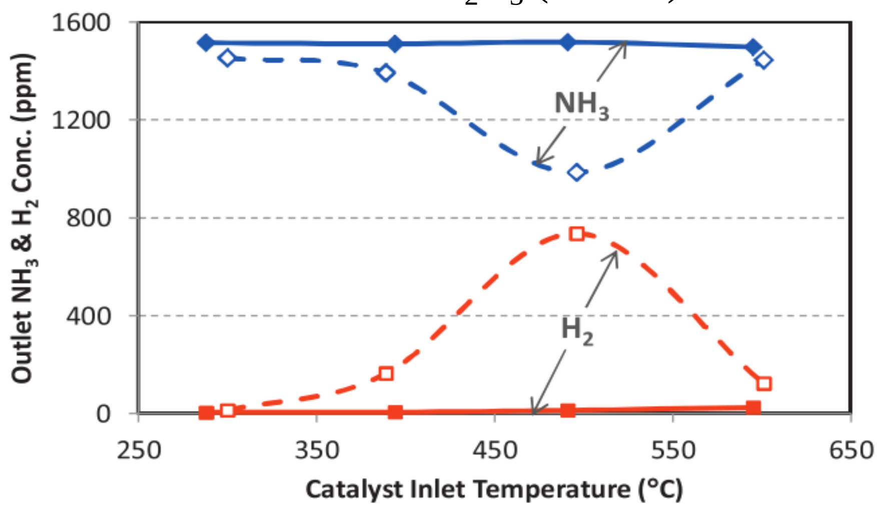


Goal: understand impact of ceria on NH_3 formation

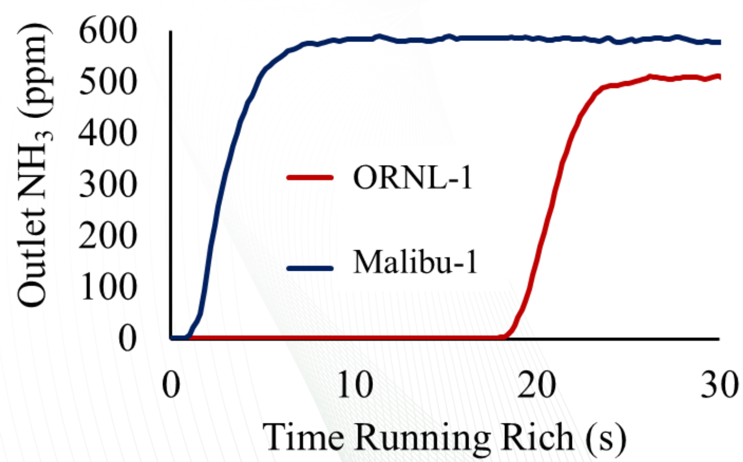
- Multiple effects of ceria*
  - Ceria promotes WGS, important for NH_3 production
  - Adding Ce and Rh promotes NH_3 decomposition
- Ceria critical for OBD/calibrations

- OSC functionality delays NH_3 production during cycling
  - ~5s breakthrough for Malibu-1 (Pd-only)
  - ~22s on ORNL-1 (OSC+NSR)

NH_3 decomposition on Pd/Al_2O_3 (solid) and $Pd/Rh/Ce/Al_2O_3$ (dashed)*



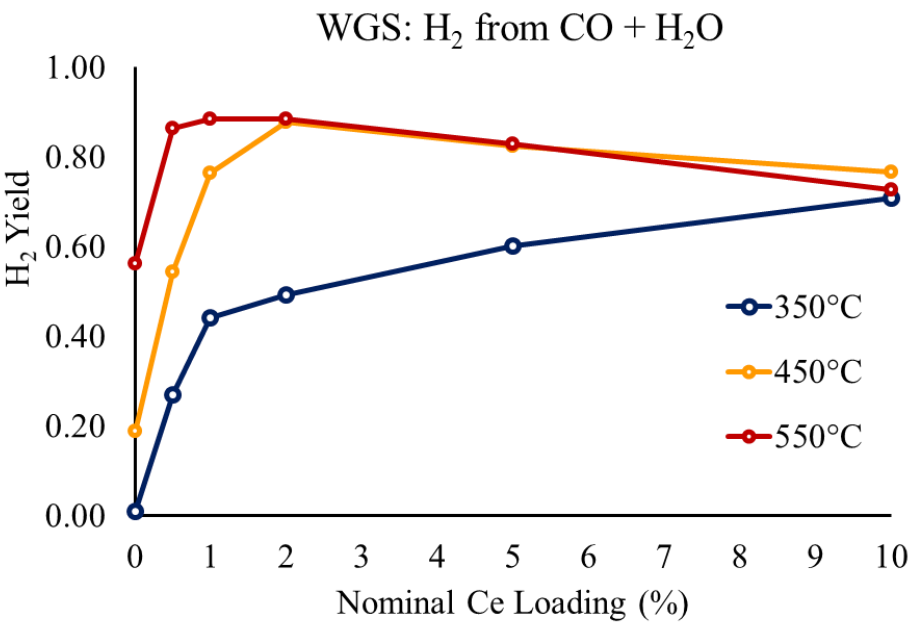
Fixed load NH_3 Production 550°C

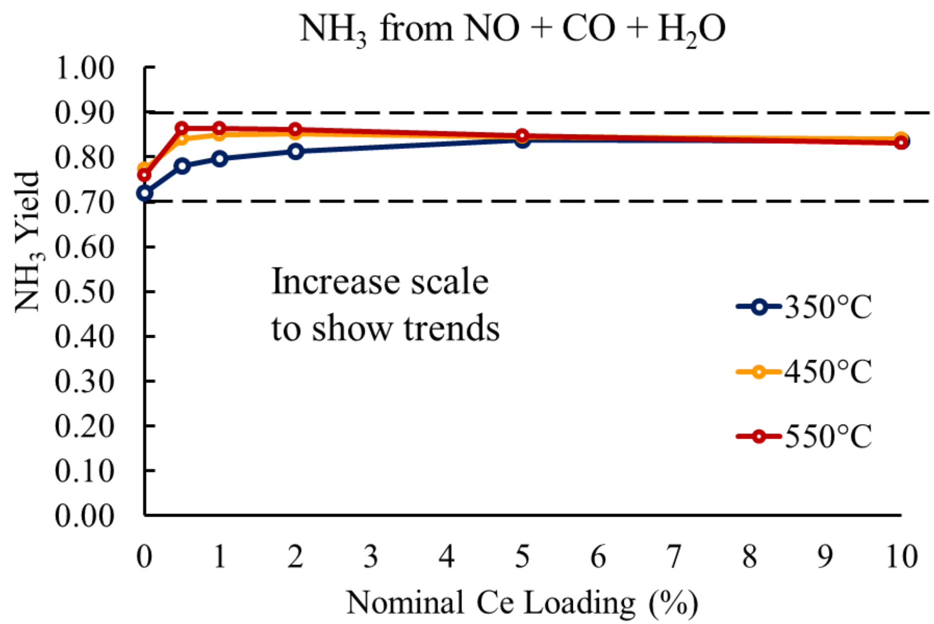


* - Oh, S. H. & Triplett, T. *Catal. Today* **231,** 22–32 (2014).

Ce is critical for low-temperature WGS reaction, but minimal Ce is necessary for formation of NH_3

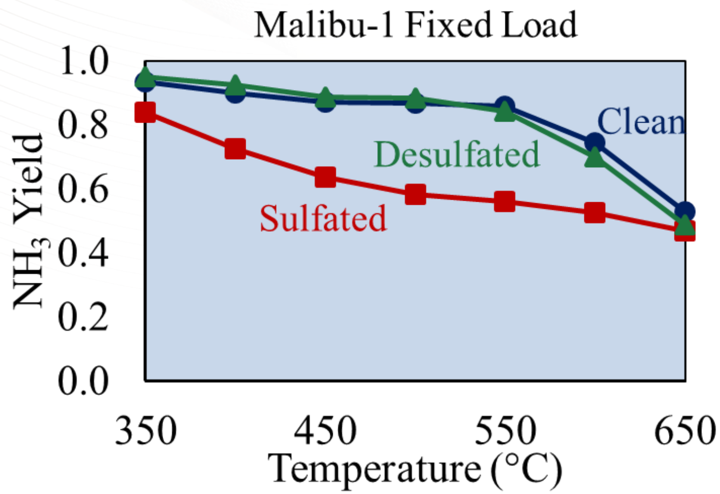
- At 350 °C, catalyst is not active for WGS without adding Ce
  - WGS (Water gas shift): $CO + H_2O \rightarrow CO_2 + H_2$
- At 350 °C, 0% Ce inactive for WGS, but still shows 70% NH_3 yield
  - Shows that formation of molecular H_2 is not necessary for formation of NH_3
  - While not necessary for NH_3 production, WGS decreases CO slip when rich

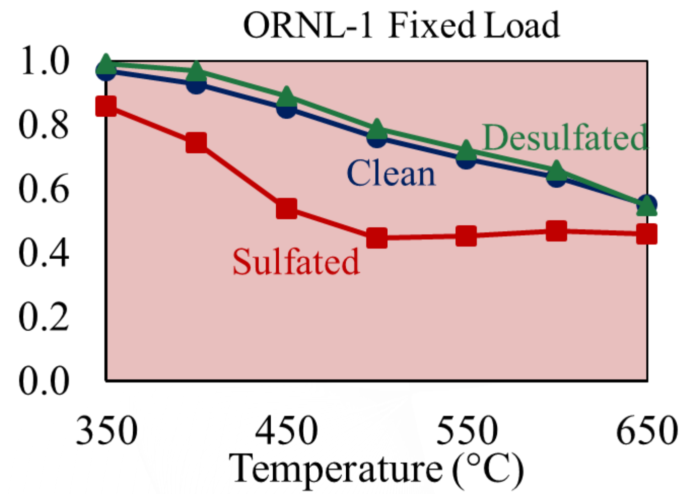




Goal: Understand impact of sulfur on NH_3 selectivity

- Both catalysts show impact of NH_3 production from sulfation
- Results difficult to deconvolute using full simulated exhaust mixture





Sulfation: 12.5 hr 2ppm SO_2 at 350°C, 1.93 $\frac{g\ SO_2}{L\ cat}$

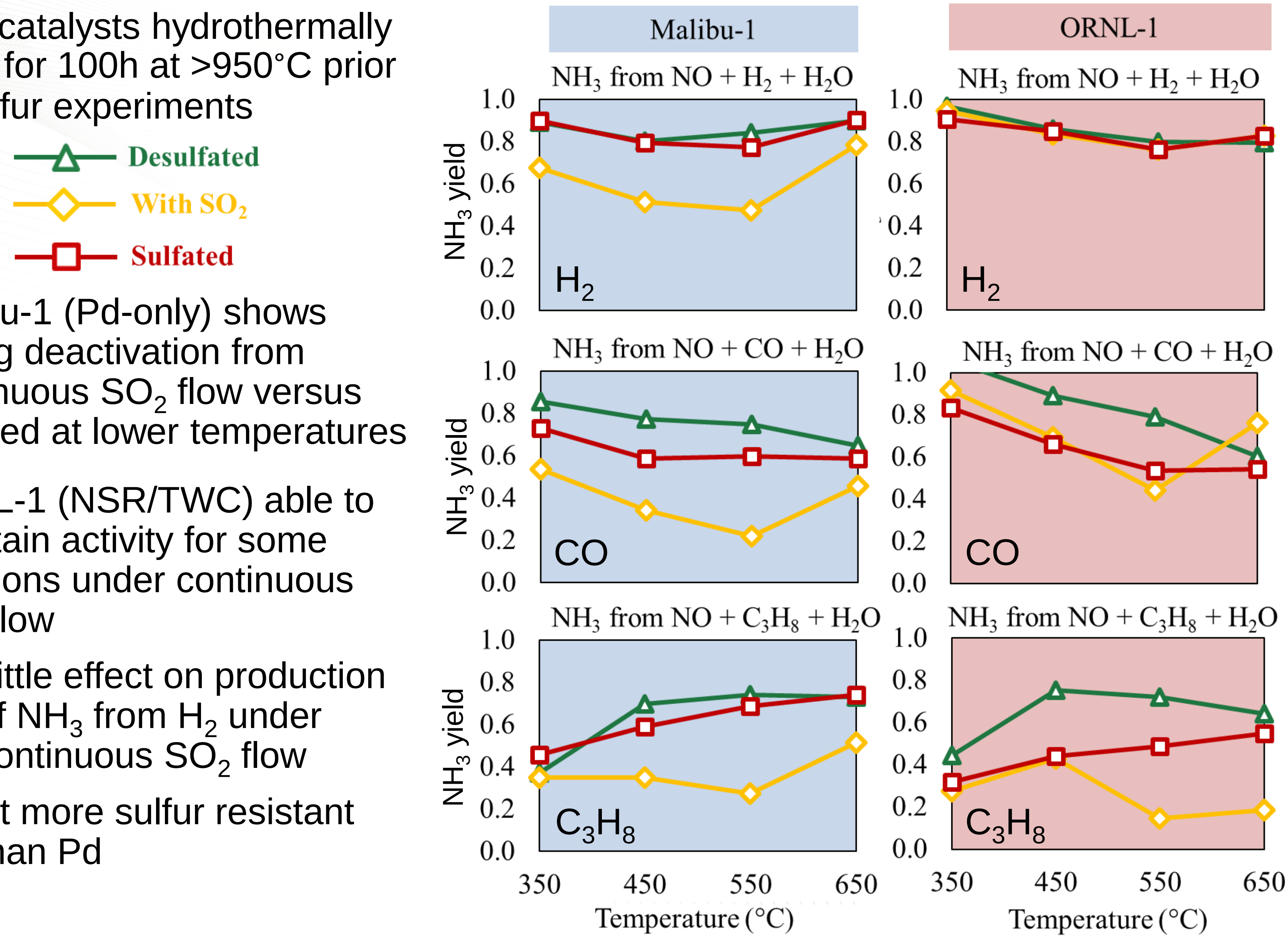
On Malibu-1: 0.438 $\frac{mol\ SO_2}{mol\ Pd}$

Desulfation at 650°C:
3 hr cycling
λ 0.97/2.0

Description	Pt g/L	Pd g/L	Rh g/L	OSC	NSC
Malibu-1	0	7.3	0	No	No
ORNL-1	2.47	4.17	0.05	Yes	Yes

S impact on NH_3 production varies w/ PGM and reductant

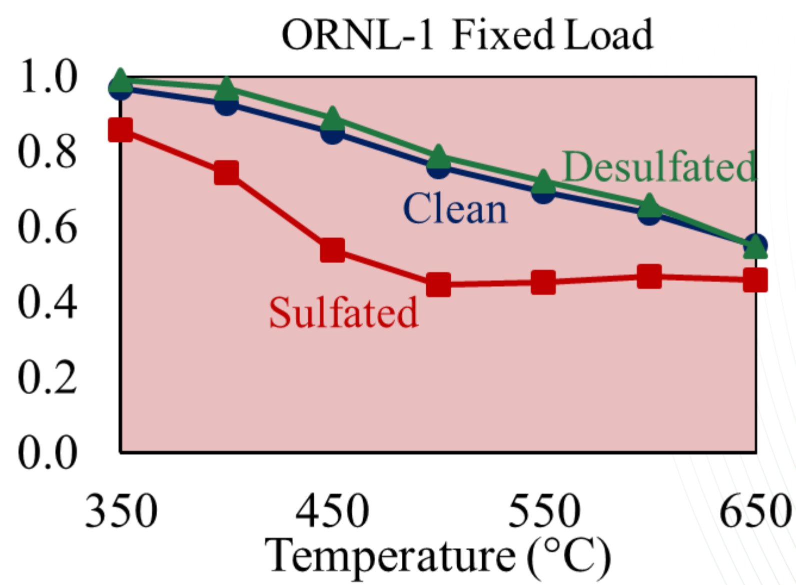
- Both catalysts hydrothermally aged for 100h at >950°C prior to sulfur experiments
- Malibu-1 (Pd-only) shows strong deactivation from continuous SO_2 flow versus sulfated at lower temperatures
- ORNL-1 (NSR/TWC) able to maintain activity for some reactions under continuous SO_2 flow
  - Little effect on production of NH_3 from H_2 under continuous SO_2 flow
  - Pt more sulfur resistant than Pd

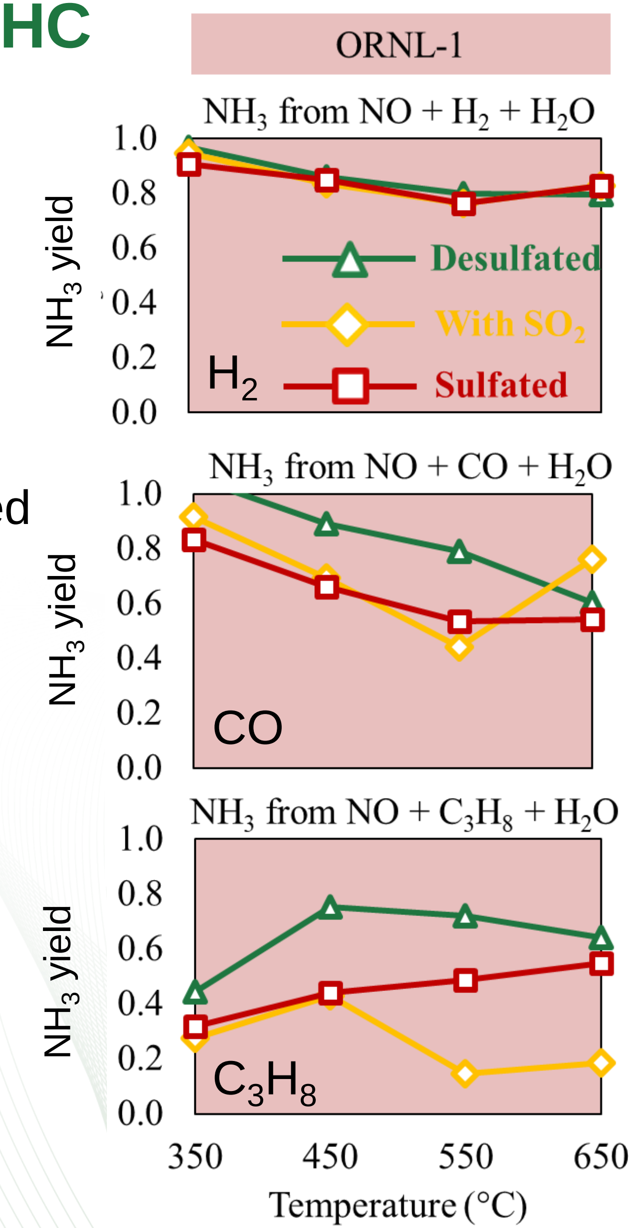


During operation, NH_3 production not only being generated through H_2; relies on CO and HC

- ORNL-1 (NSR/TWC) able to maintain NH_3 production activity while flowing H_2 under continuous SO_2 flow
  - NH_3 production from CO and C_3H_8 impacted while flowing SO_2 and after sulfating
- Under simulated exhaust conditions (below), NH_3 production on the sulfated catalyst is clearly impacted
  - Indicates CO and HCs pathways to NH_3 are followed

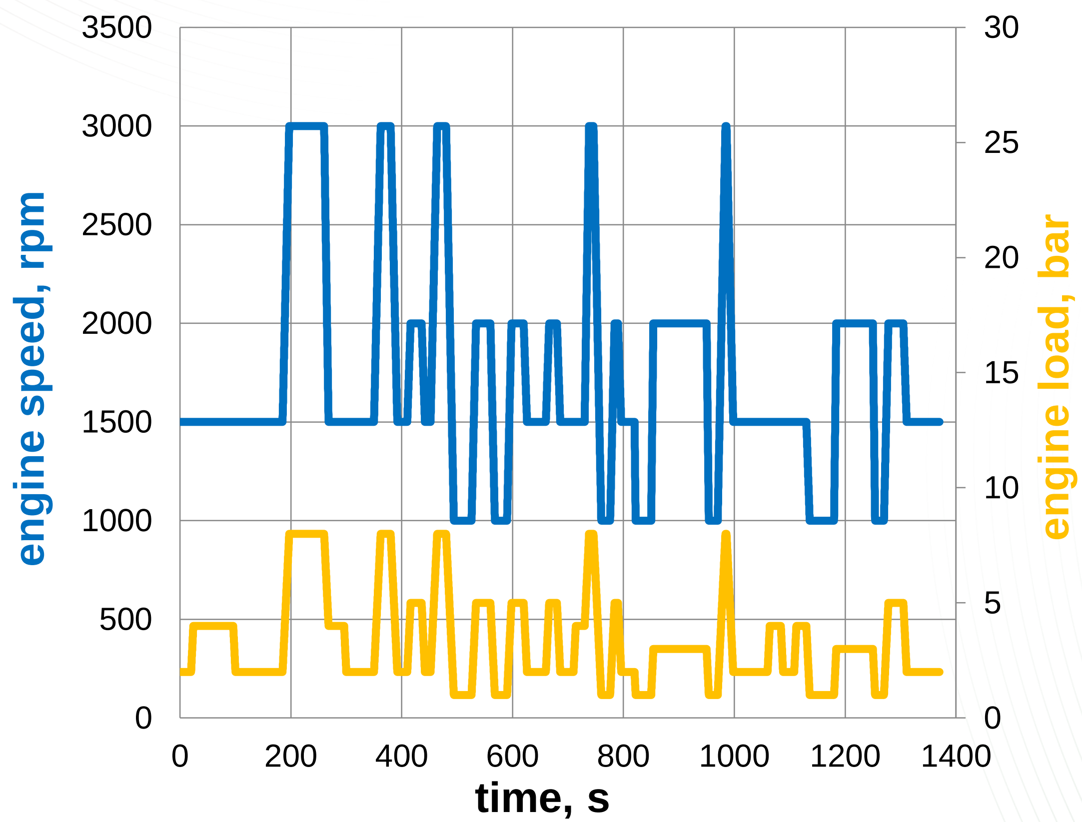
NH_3 from NO + H_2 + CO + C_3H_8 + O_2





To simulate drive cycle, GM provided 6-mode pseudo-transient cycle utilized for passive SCR evaluation

- Operating pseudo-transient cycle closely captures fuel consumption benefit relative to stoichiometric observed on vehicle in study*
  - 9.6% with pseudo-transient drive cycle
  - 10% with FTP vehicle study



6 speed/load modes

Speed [rpm]	Load [bar]	Default Mode
1000	1.0	LS
1500	2.0	LS
1500	4.0	LS
2000	3.0	LS
2000	5.0	LH
3000	8.0	Stoich

LS=lean stratified, LH=lean homogeneous

* - SAE2010-01-2267, SAE2011-01-1218

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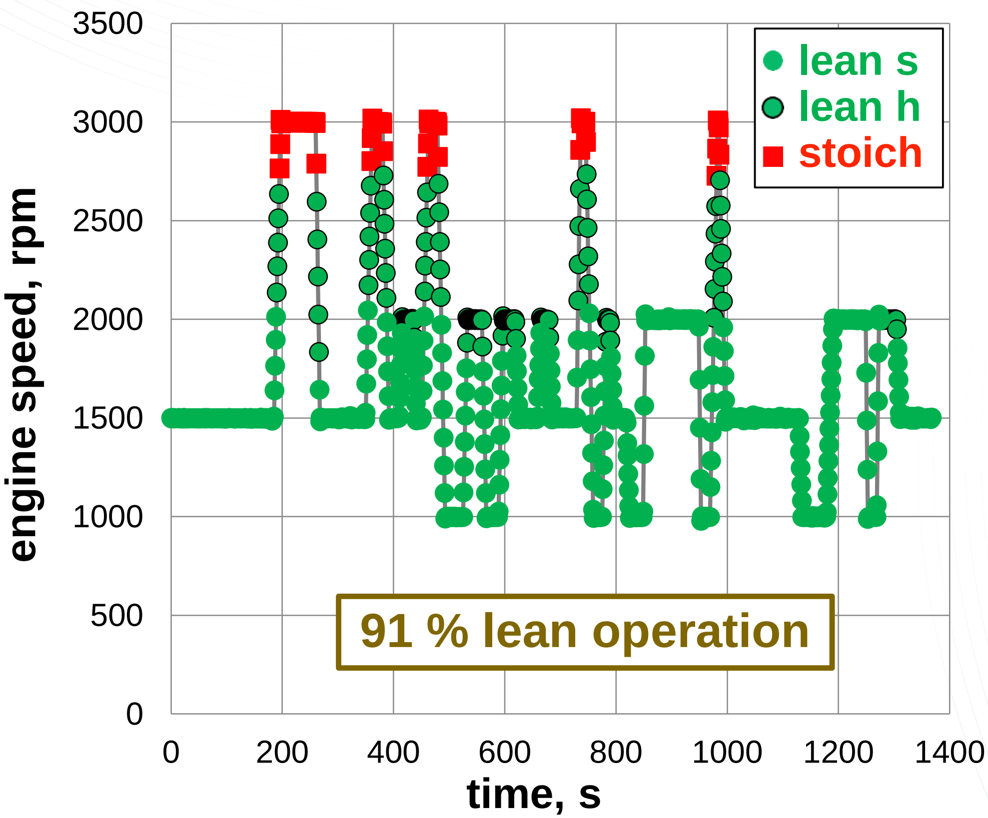

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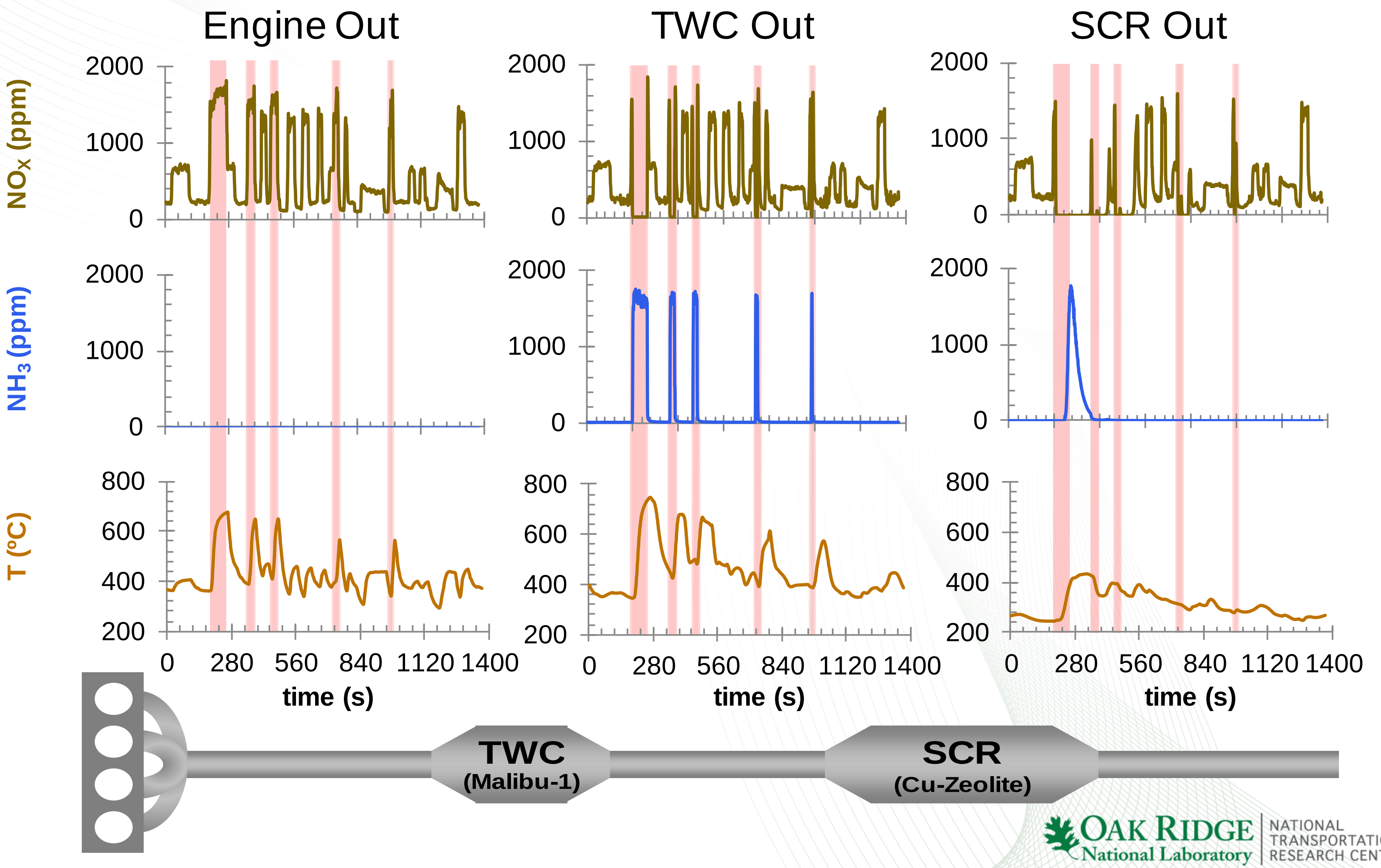
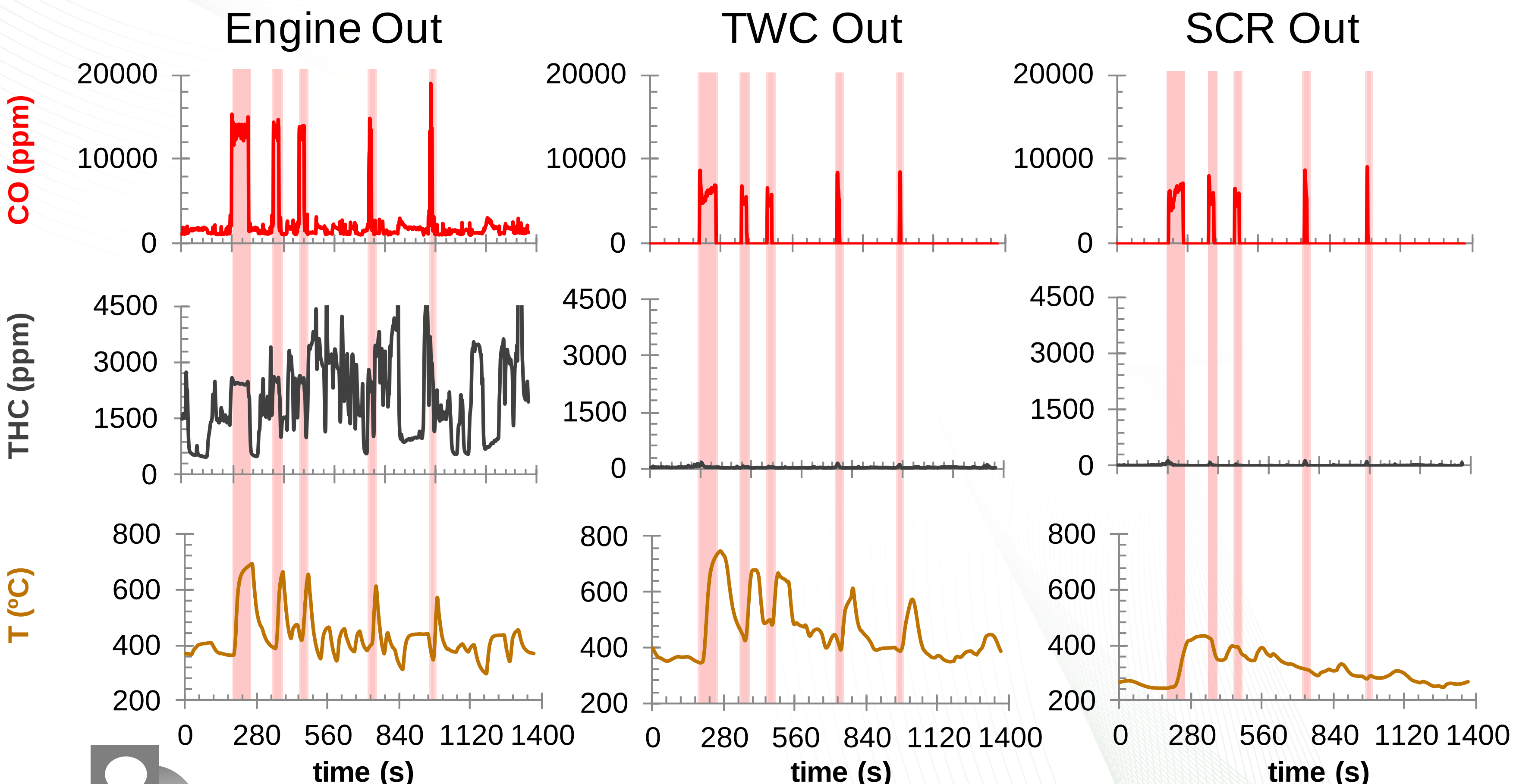
Initial operating strategy: operate at $\lambda = 0.96$ instead of 1.0

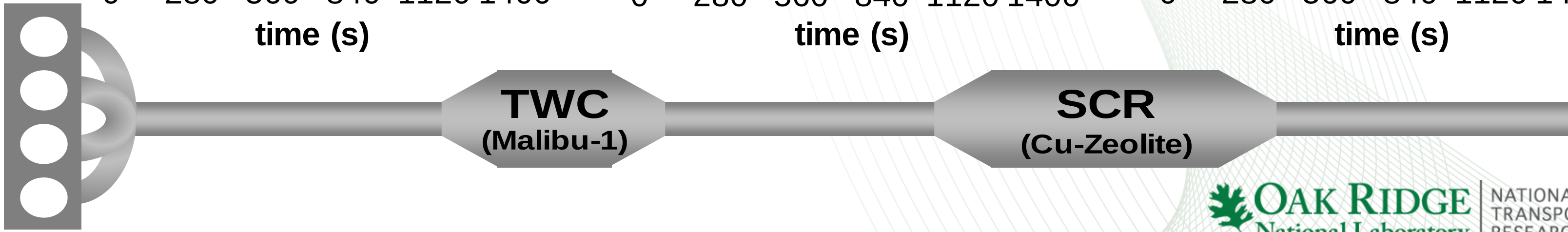
- Only substituting rich operation for stoichiometric points does not generate enough NH_3
- Hard acceleration results in high engine out NO_X flux and TWC NH_3 production, but high temperatures that accompany hard acceleration prevent NH_3 storage on SCR



Initial operating strategy: operate at λ = 0.96 instead of 1.0

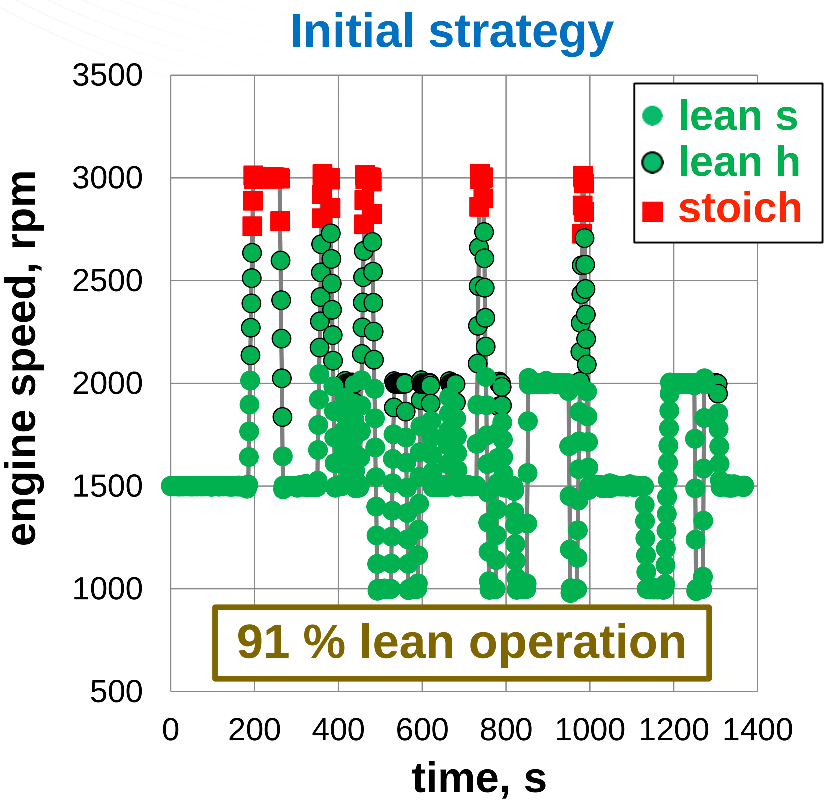
- Significant HC reduction under both rich and lean conditions
- High CO slip during rich operation

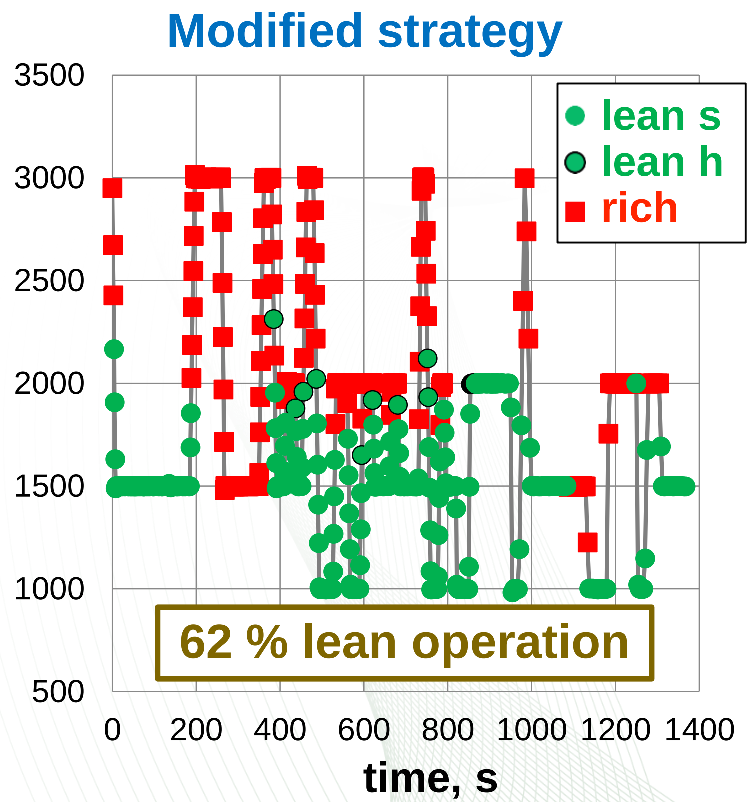




Modified operating strategy for better NOx control

- Partially preload SCR with NH_3 to have enough for first 200s
- Operate at λ=0.97 instead of stoichiometric under most conditions
  - when SCR temperature too high to store NH_3, operate λ=0.99
- Operate rich instead of lean homogeneous
- Switch to rich if NOx slip > 10ppm

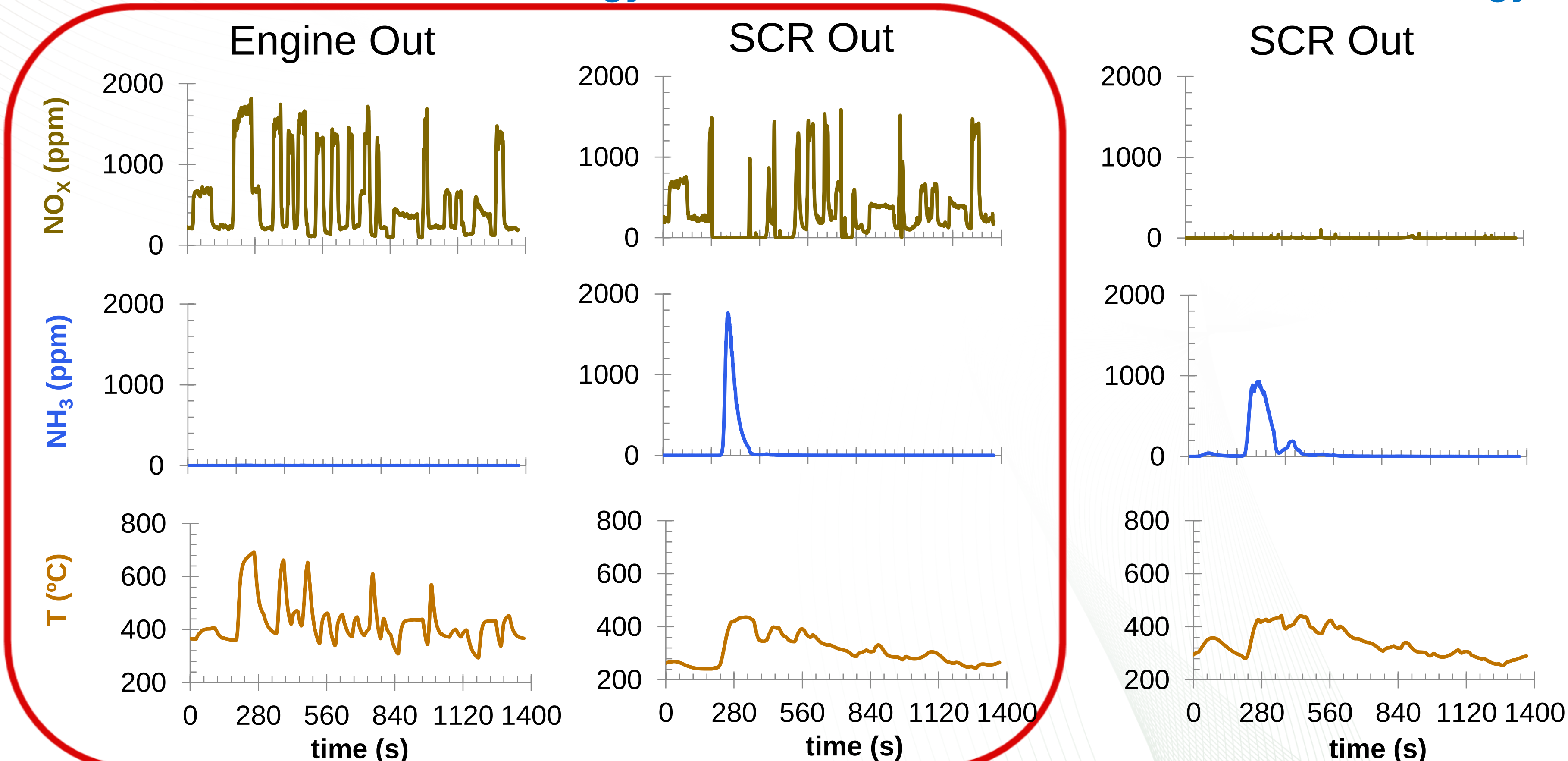


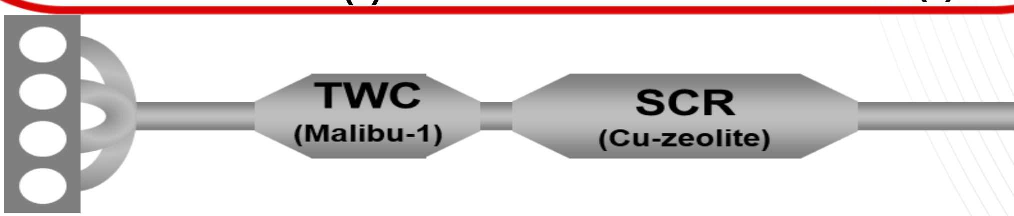


Modified operating strategy for better NOx control

- NOx essentially eliminated
- NH_3 slip still observed, indicating improved fuel efficiency possible

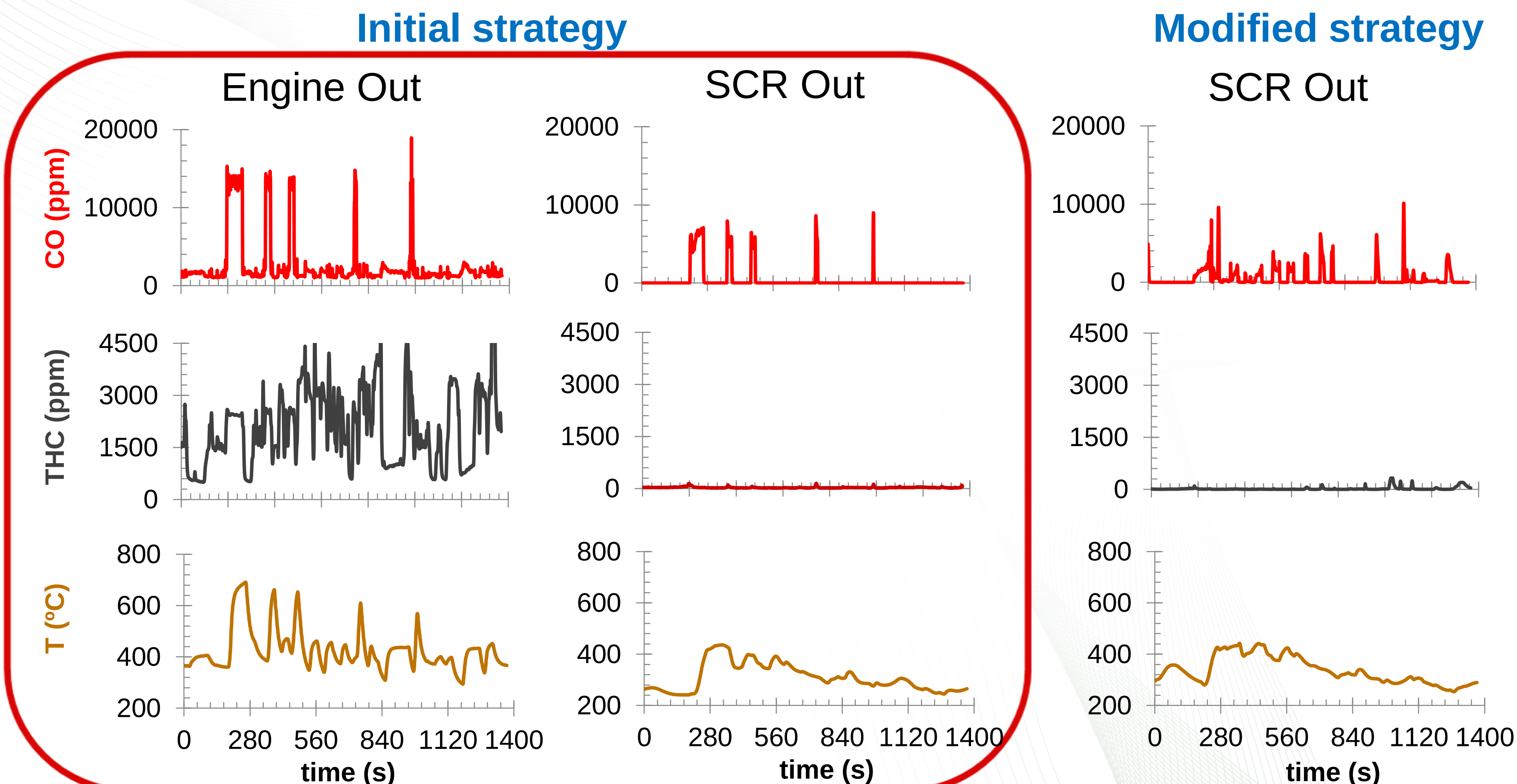
Initial strategy | Modified strategy

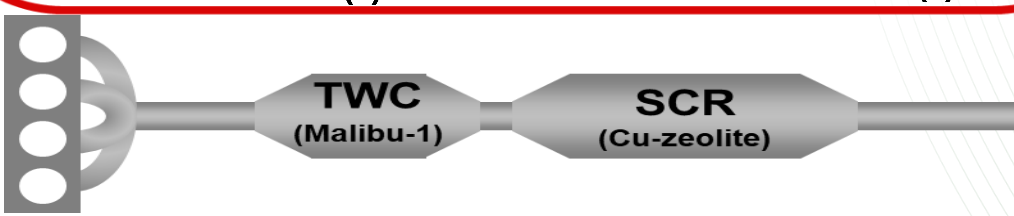




Modified operating strategy for better NOx control

- CO slip is still significant; points to need for clean-up catalyst
- HC slip is still very low, but very close to the NOx+HC emission limit





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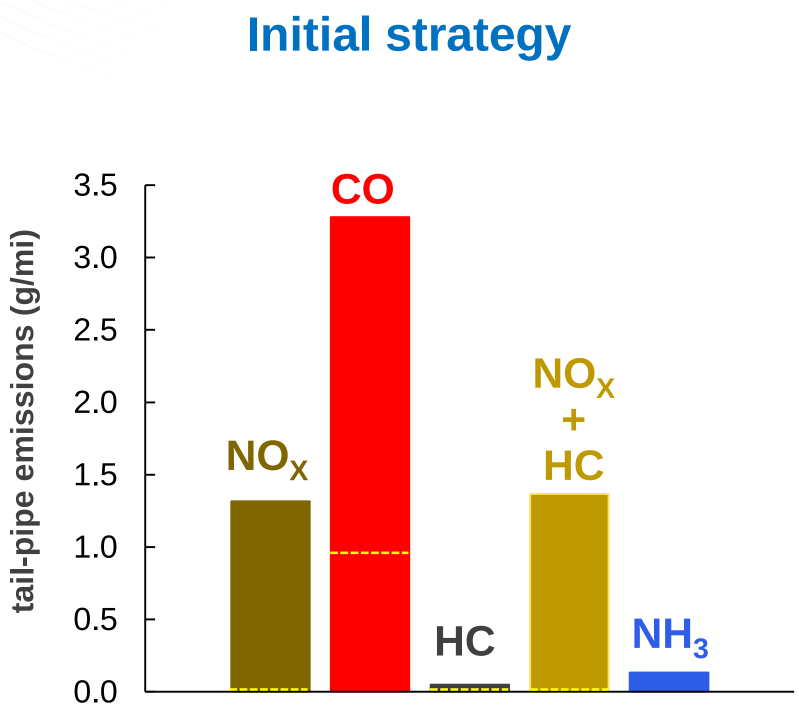
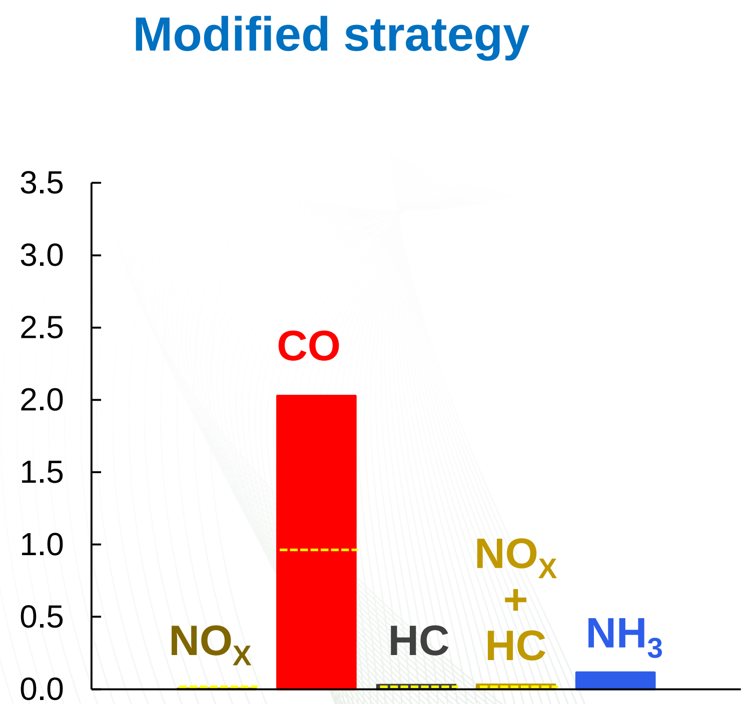
Improved operation strategy illustrates potential of passive SCR, as well as challenges and opportunities

- Achieved 5.9% fuel efficiency improvement with 0.03 g/mi NOx + HC
  - NH_3 slip indicates that additional fuel efficiency can be gained
- CO twice emission standard but with a high OSC clean-up catalyst it can likely be achieved through oxidation and WGS





------ Tier 3 bin 30: 0.03 g/mi of NOx+HC, 1.0 g/mi of CO

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Remaining Challenges

- Need more good engineers/ scientists in work force
- CO and HC control during rich conditions
- Maximize fuel efficiency while maintaining or further reducing emissions

Future Work*

- Calvin Thomas expected to receive PhD in Summer/Fall 2018
- Experiments planned on flow reactor and engine platform to evaluate impact of clean-up catalyst
  – Includes potential addition of secondary air
- Engine evaluation of efficiency optimized emissions control system on pseudo-transient cycle
  – Including temperature control for NH_3 storage
  – Additional catalyst technologies

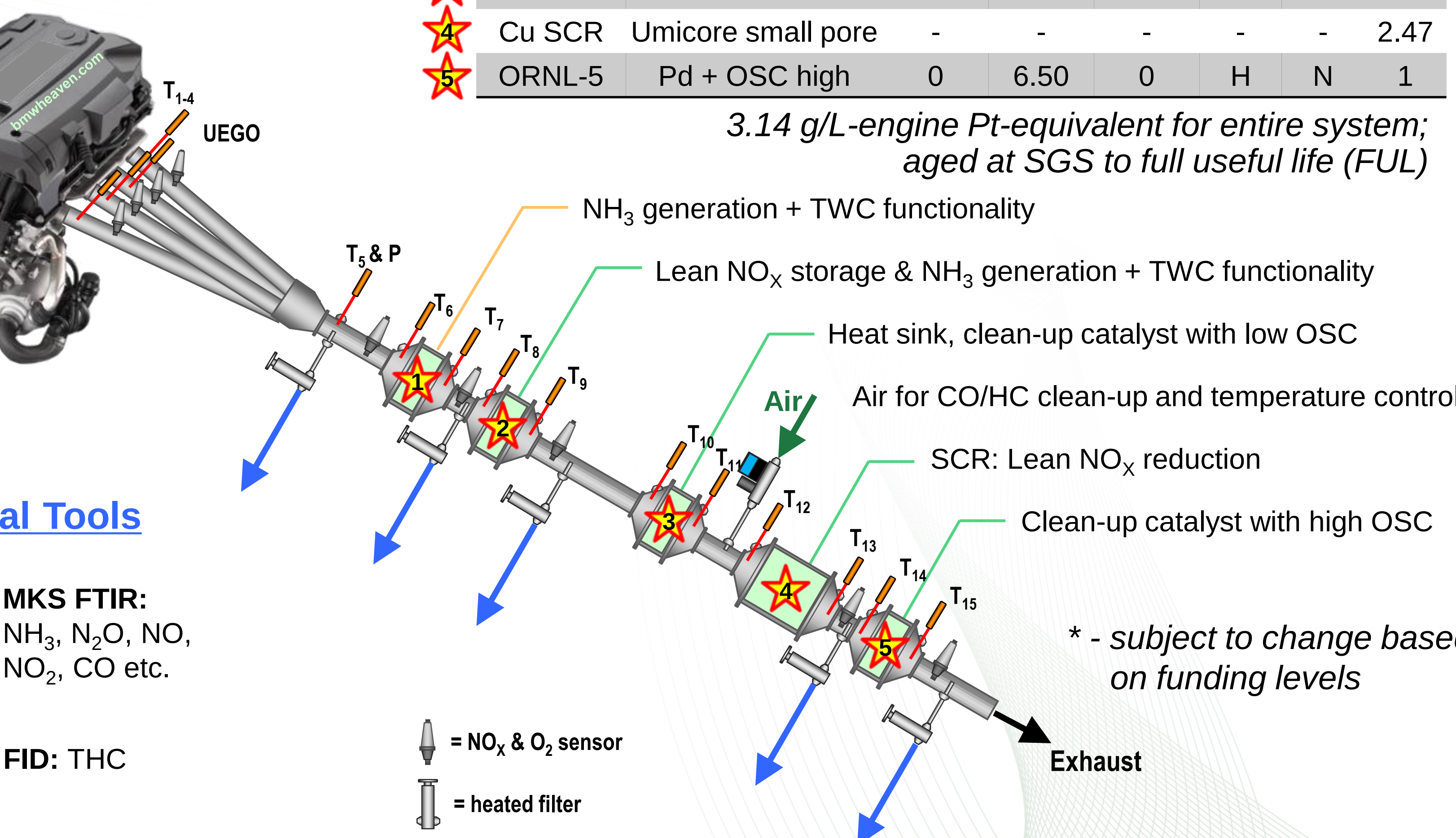
** - subject to change based on funding levels*

Future work: evaluate passive SCR system architecture for maximum fuel savings while meeting Tier 3 NOx+HC/CO*

BMW 120i 4-cylinder 2.0-liter naturally aspirated lean gasoline engine platform with National Instrument open controller

	sample ID	Description	Pt (g/l)	Pd (g/l)	Rh (g/l)	OSC	NSC	Vol (l)
1	Malibu-1	Front half of TWC	0	7.3	0	N	N	0.65
2	ORNL-1	Pt + Pd + Rh	2.47	4.17	0.05	Y	Y	0.65
3	ORNL-3	Pd + OSC low	0	0-1.4	0	L	N	1
4	Cu SCR	Umicore small pore	-	-	-	-	-	2.47
5	ORNL-5	Pd + OSC high	0	6.50	0	H	N	1

3.14 g/L-engine Pt-equivalent for entire system; aged at SGS to full useful life (FUL)



** - subject to change based on funding levels*

Analytical Tools

MKS FTIR: NH_3, N_2O, NO, NO_2, CO etc.

FID: THC

SpaciMS: H_2 & O_2

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Responses to 2017 4 Reviewers (overall 3.3/4)

Summary of Reviewers' Feedback:

1. Lots of positive feedback across all categories, particularly:
   a) Well-integrated innovative approach
   b) Transient approach is well-thought out
   c) Excellent example leveraging knowledge
   d) Well-coordinated, relevant research
2. Need to assess the emissions over a Federal Test Procedure (FTP) cycle including challenging speed/load points
3. Aging of the SCR under conditions other than high temperature
4. Costs of sensors, additional non-PFM components, controls, OBD should be considered
5. Interest in how excess NH_3 translates into tailpipe NOx emissions
6. Keep an eye on N_2O formation
7. What to do when engine at end-of-life?

Project Responses:

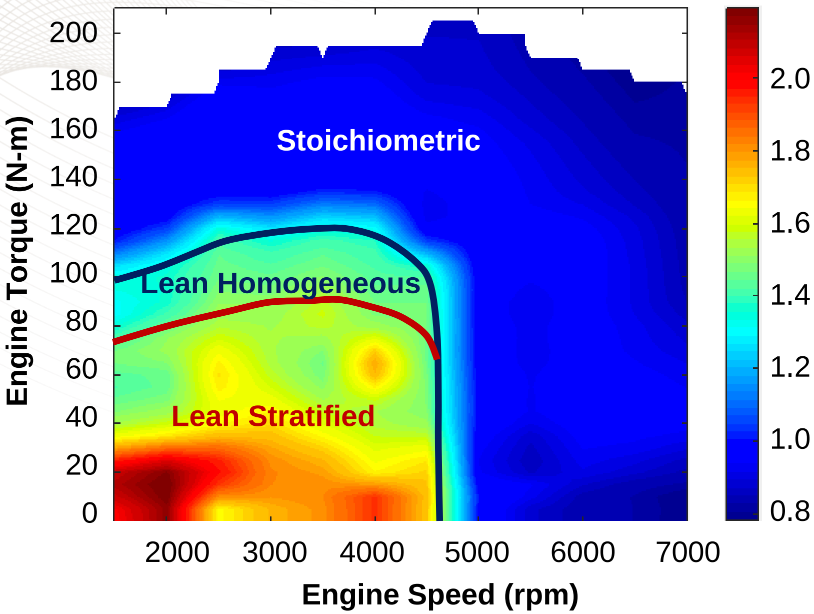
1. THANKS!
2. Transient drive cycle implemented this year to address this concern
3. Have sent a series of TWCs to SGS for 4-mode aging; plans for SCR too
   a) minimal impact observed to date after 100+ hour operation on engine
4. Cost is a consideration of this project, but not appropriate for us to assign costs to these components
5. Tracking currently and in future; data does not suggest re-oxidation to NOx
6. Doing this; data in backup slides
7. Have plans for future engine platform

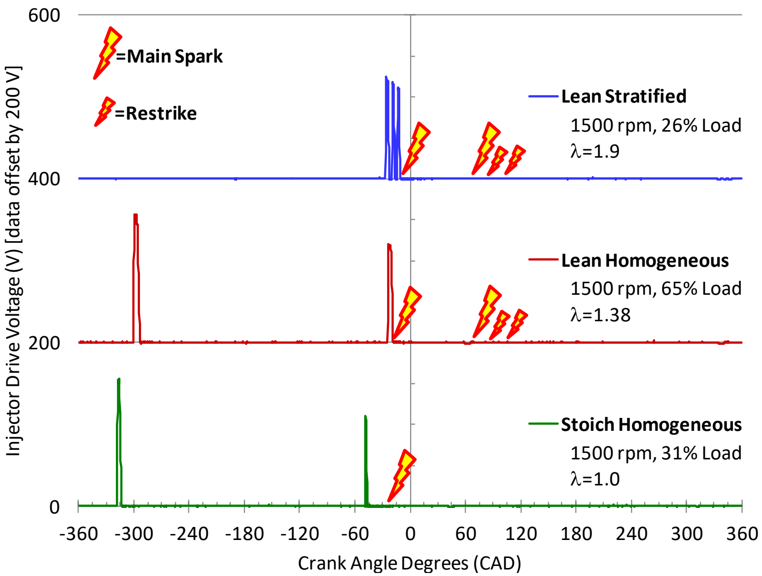
Summary

- **Relevance**
  - Lean GDI engine emission control enables potential 10-15% fuel efficiency gain for gasoline-dominant U.S. light-duty fleet
- **Approach**
  - Bench flow reactor, engine, and aging studies are combined to study fuel efficiency and emissions relative to Tier 3 standard
- **Technical Accomplishments**
  - Full Review of project at USCAR in August 2017
  - Evaluated impact of Ce loading on NH_3 production over model TWC catalyst formulations
  - Completed sulfur sensitivity analysis on two TWCs
  - Using two catalyst system met Tier 3 NOx+HC (0.03 g/mi) with 5.9% fuel efficiency improvement
- **Collaborations**
  - GM, Umicore, and the University of South Carolina are primary partners
- **Future Work** *(subject to change based on funding levels)*
  - Calvin Thomas to receive PhD in Summer/Fall 2018
  - Flow reactor and engine platform to evaluate impact of clean-up catalyst
  - Evaluation of efficiency optimized emissions control system on pseudo-transient cycle

Technical Back-Up Slides

BMW 120i engine features three main combustion modes





- Center mounted combustion system design
- Lean Stratified
  - fuel injections close to TDC
  - multiple spark events
  - lambda ranges between 1.6 and 2.2
  - limited to 4500 rpm and 55% load

- Lean Homogeneous
  - two injections: one during intake stroke and one late in compression stroke close to TDC
  - multiple spark events
  - λ ranges between 1.4 and 1.6
  - limited to 4500 rpm and 55-75% load
- Stoichiometric
  - two injections: one during intake stroke and a smaller one early in compression stroke
  - single spark event
  - λ=1
  - entire engine operating range

Three-Way Catalyst (TWC) Sample Matrix

- **"Malibu" TWCs:**
  - Commercial state-of-the-art TWC from a MY2009 Chevrolet Malibu SULEV vehicle
- **"ORNL" TWCs:**
  - Prototype formulations supplied by Umicore specifically for this project

Catalyst Sample Matrix [OSC=oxygen storage capacity; NSC=NOx storage capacity]

sample ID	Description	Pt (g/l)	Pd (g/l)	Rh (g/l)	OSC	NSC
Malibu-1	Front half of TWC	0	7.3	0	N	N
Malibu-2	Rear half of TWC	0	1.1	0.3	Y	N
Malibu-combo	Full TWC	0	4.0	0.16	Y	N
ORNL-1	Pt + Pd + Rh	2.47	4.17	0.05	Y	Y
ORNL-2	Pd + Rh	0	6.36	0.14	N	N
ORNL-6	Pd	0	6.50	0	N	N
ORNL-5	Pd + OSC high	0	6.50	0	H	N
ORNL-4	Pd + OSC med	0	4.06	0	M	N
ORNL-3	Pd + OSC low	0	1.41	0	L	N

Cycling flow reactor experiments to estimate TWC effects on fuel consumption and mimic portions of FTP

- Used feedback-controlled cycles on flow reactor to evaluate dynamic TWC response in context of passive SCR
- Evaluated two different simulated engine cycles (fixed load, load step)

	fixed load		load step	
	rich	lean	rich	lean
load (BMEP)	2 bar	2 bar	8 bar	2 bar
SV (h^{-1})	**27000**	45000	**60000**	45000
NOx (ppm)	**600**	360	**1200**	360
max lean time	**50%**		**80%**	
simulates	cruise		“hill” transient	

	Rich						Lean
λ	0.95	0.96	0.97	0.98	0.99	1.00	2
O_2 (%)	0.96	1.02	1.07	1.13	1.17	1.22	10
CO (%)	2.0	1.8	1.6	1.4	1.2	1.0	0.2
H_2 (%)	1.0	0.9	0.8	0.7	0.6	0.5	0
NO (ppm)	600 (or 1200)						360
C_3H_8 (ppm C_1)	3000						1900
H_2O (%)	11						6.6
CO_2 (%)	11						6.6
TWC SV (hr^{-1})	27000 (or 60000)						45000

- Compositions & flows selected to mimic BMW GDI engine exhaust
- Space velocity changed with λ and load
- C_3H_8 chosen as challenging HC

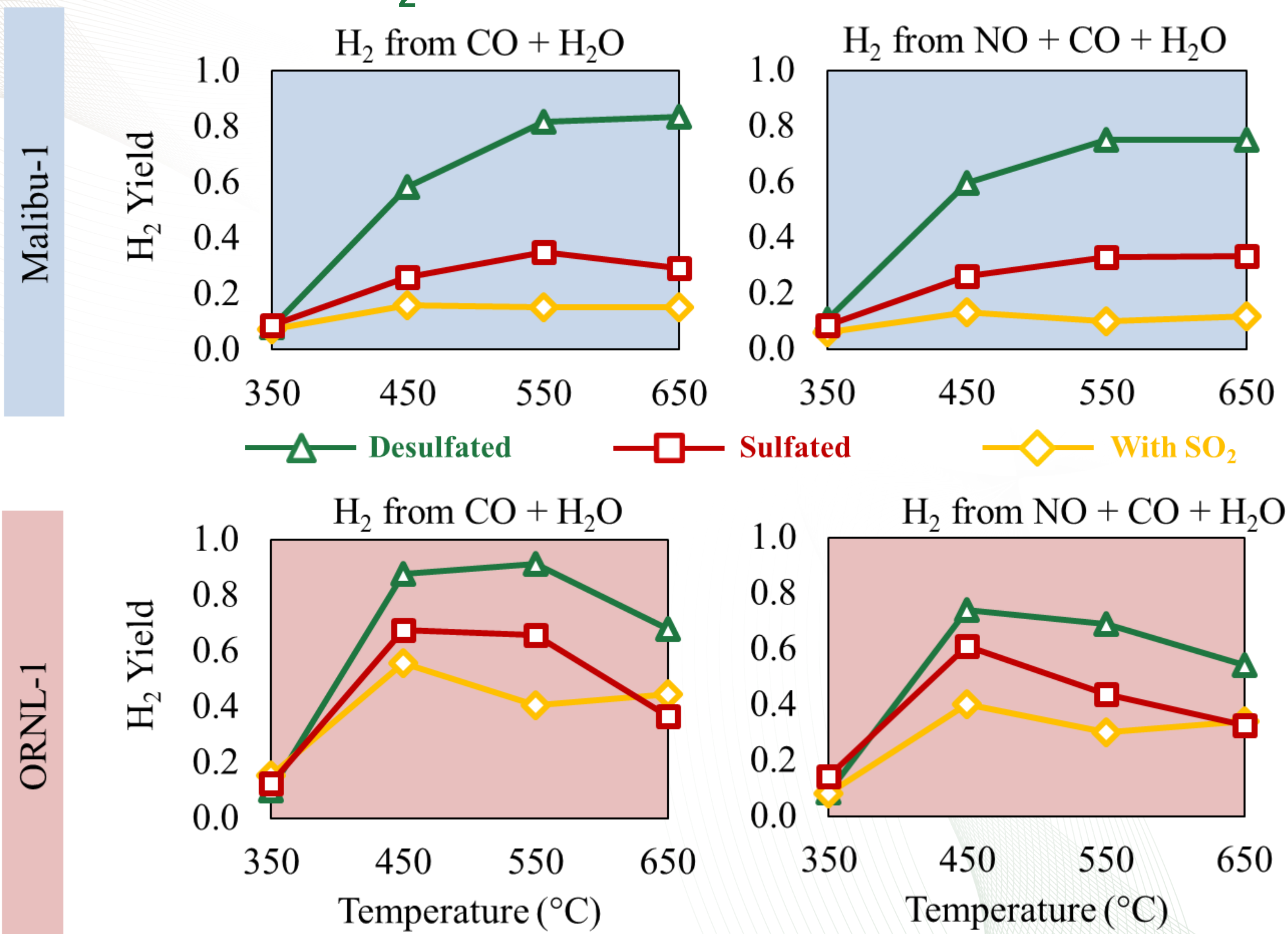
Employing isolated reactions to understand S impact

- Catalysts evaluated in multiple states:
  - desulfated, sulfated, and with SO_2 in stream
- 2 min lean – 2 min rich, no feedback control
- Cycled between 10% O_2 and N_2 balance with SV = 27,000 hr $^{-1}$

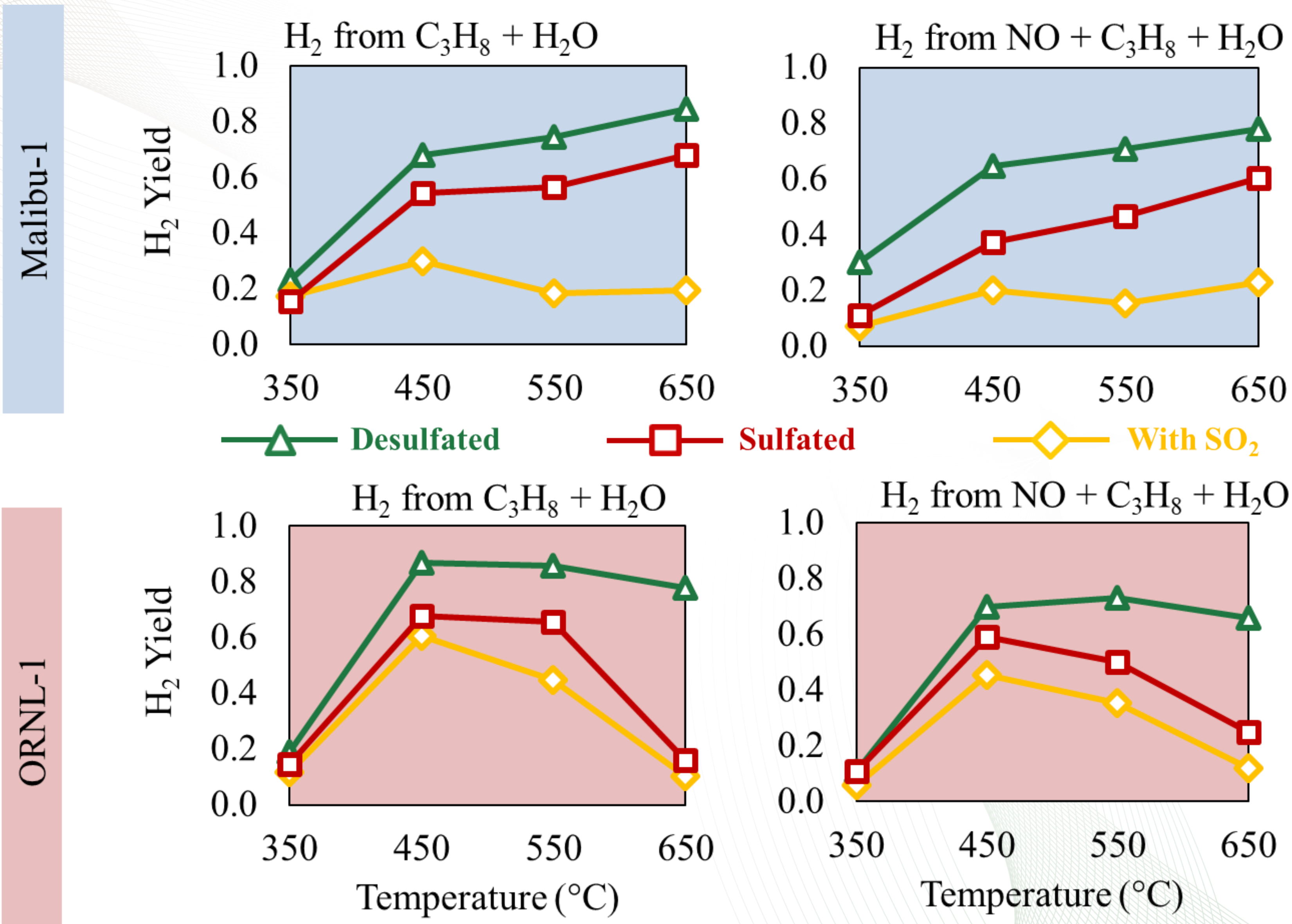
	NH_3 from H_2 $NO + H_2 + H_2O$		NH_3 from CO $NO + CO + H_2O$		NH_3 from C_3H_8 $NO + C_3H_8 + H_2O$		WGS $CO + H_2O$		Reforming $C_3H_8 + H_2O$	
	Rich	Lean	Rich	Lean	Rich	Lean	Rich	Lean	Rich	Lean
CO (%)	0	0	1.0	1.0	0	0	1.0	1.0	0	0
H_2 (%)	1.0	1.0	0	0	0	0	0	0	0	0
NO (%)	0.05	0.05	0.05	0.05	0.05	0.05	0	0	0	0
C_3H_8 (%)	0	0	0	0	0.1	0.1	0	0	0.1	0.1
H_2O (%)	5.0	5.0	5.0	5.0	5.0	5.0	5.0	5.0	5.0	5.0
O_2 (%)	0	10.0	0	10.0	0	10.0	0	10.0	0	10.0

Description	Pt g/L	Pd g/L	Rh g/L	OSC	NSC
Malibu-1	0	7.3	0	No	No
ORNL-1	2.47	4.17	0.05	Yes	Yes

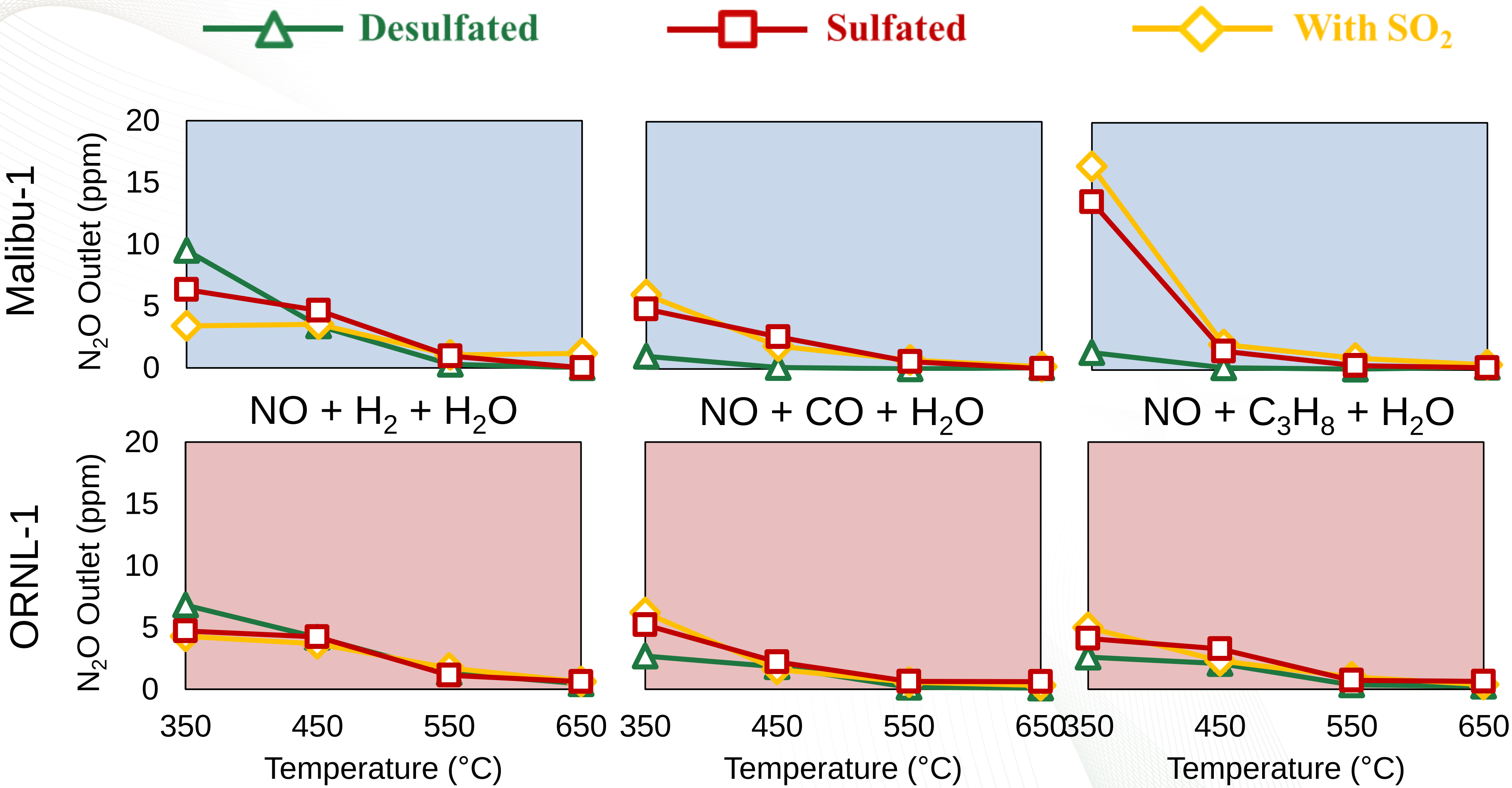
H_2 Production from CO



H_2 Production from C_3H_8

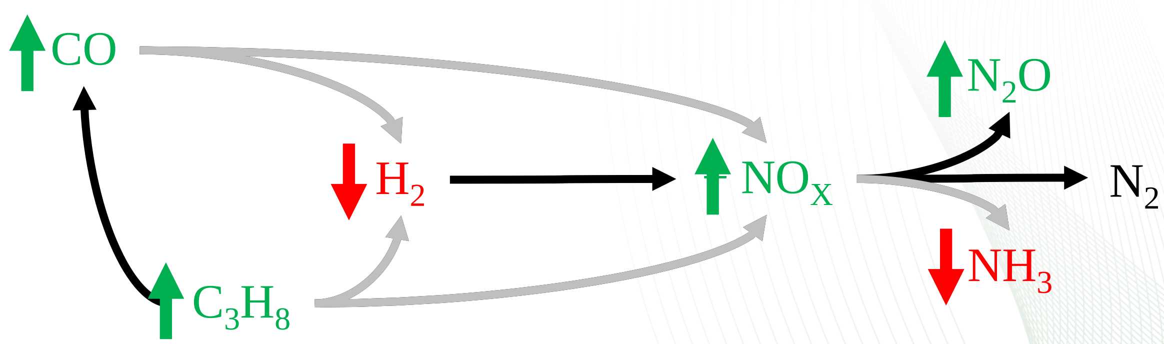


S impact on N_2O production in isolated reactions



Observed impact of sulfations on passive SCR chemistry

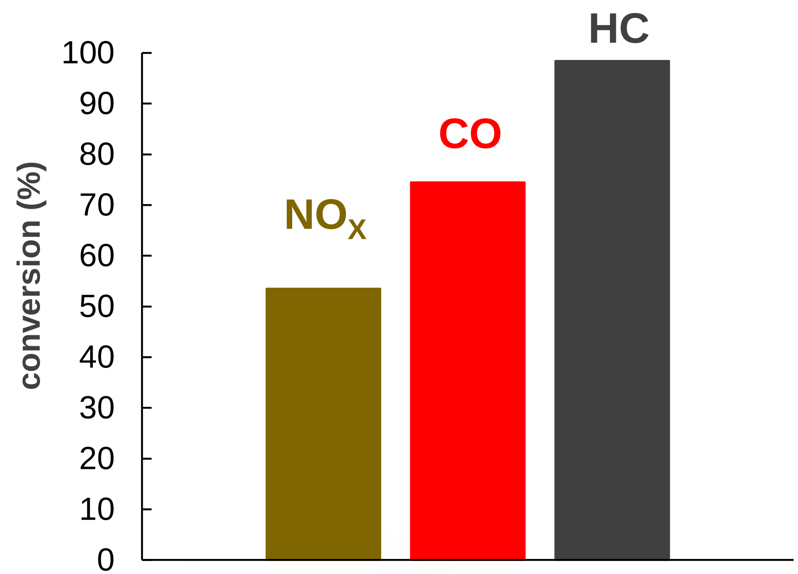
- Before sulfation:
  - CO and C_3H_8 contribute to formation of H_2
  - Reductants can be oxidized to form H_2O
  - Reaction with NO_X to form NH_3 is preferred
- Effects of sulfation on isolated reactions:
  - Production of H_2 and NH_3 from CO and C_3H_8 deactivated
  - Significant N_2O formation
- Isolated reactions account for changes in simulated exhaust:
  - Increased CO, C_3H_8, NO_X, N_2O
  - Decreased H_2, NH_3

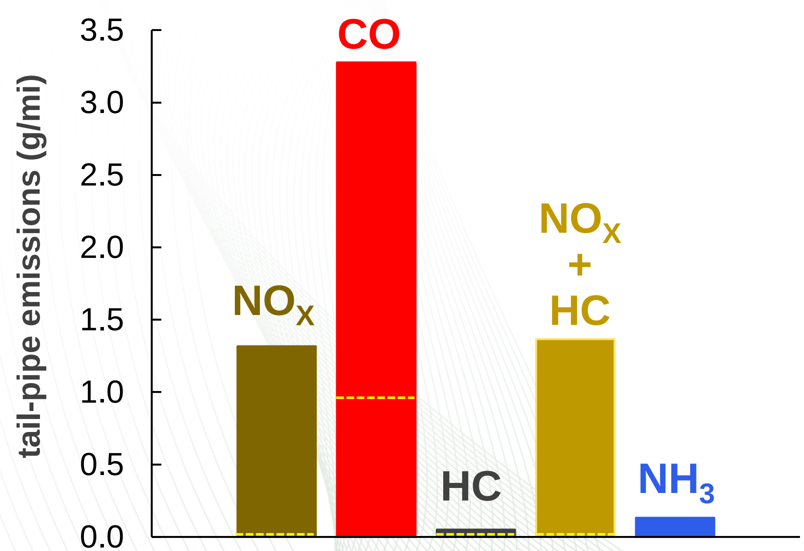


Initial strategy shows challenges and opportunities for passive SCR system

- 1% fuel penalty for running λ=0.96 vs. 1.0
- NOx conversion
  - Low NOx conversion 54%
  - Cumulative engine-out lean NOx>rich NOx
    - Not enough NH_3 to treat lean NOx
    - Possible solution: run rich more often, or store lean NOx (TWC w/ NOx storage, future direction)
    - Transitional lean homogeneous mode generates a lot of NOx, 15% of lean ran in lean homogeneous but generated 53% of lean NOx → run stoich instead
- CO emissions challenging without clean-up catalyst
- NH_3 slip
  - Opportunity for better fuel efficiency
  - Need for better SCR temperature control

------ Tier 3 bin 30: 0.03 g/mi of NOx+HC
1.0 g/mi of CO





Pt-equivalent calculation basis

	5-year Average ($/troy oz.)	Pt-equivalent
Platinum	$ 1,504/troy oz.	1.0
Palladium	$ 463/troy oz.	0.3
Rhodium	$ 3,582/troy oz.	2.4
Gold	$ 989/troy oz.	0.7

* - will use Pt equivalent cost to account for different costs of Pt, Pd and Rh; 5-year average value fixed at beginning of project

As a reference point, the BMW 120i vehicle with a Euro 5 compliant TWC+LNT system contains a Pt-equivalent total of 5.1 g/liter of engine displacement